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This research program developed new understanding into the fundamental nature of nonlinear laser spectroscopy of resonant systems in the presence of state specific reservoir coupling. Experimental studies to support this work were performed in atomic sodium.

However, the results of this work were extended to more complex systems. In particular, we demonstrated that these advanced nonlinear laser spectroscopy methods could be applied to obtain new understanding in many body systems such as GaAs quantum well structures. For example, one of the most interesting achievements of this program was the first direct observation of the homogeneous line shape of the HHL exciton in a GaAs/AlGaAs multiple quantum well. Another interesting result of this work was the clear demonstration of the nature of exciton spectral diffusion.

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NON LINEAR OPTICAL STUDIES OF RESONANT SYSTEMS

FINAL REPORT

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**Final Report to U.S. Army Research Office**

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**Nonlinear Optical Studies of Resonant Systems**

The objective of this program has been to develop a fundamental understanding of the four-wave mixing spectral response in resonant systems and apply this understanding to semiconductor heterostructures. A summary of our current progress shows:

Demonstration of narrow resonances in the resonant nonlinear response from noncancellation of quantum mechanical amplitudes due to nonconservation of population, alignment and orientation.

Demonstration of collision induced narrow resonances due to state changing and velocity changing collisions.

Demonstration of Raman type transitions to the nonlinear response, characterized by widths determined by the Raman coherence.

Study of high optical field effects in three level systems.

Measurements of phonon mediated relaxation by FWM in semiconductor doped glass.

Use of FWM for measurement of excitation rates at low temperature in GaAs quantum well structures. Demonstration of multiple state contributions.

Use of FWM to eliminate inhomogeneous broadening at low temperature to obtain a direct measurement of the homogeneous lineshape of the heavy hole exciton in GaAs quantum well structures.

Demonstration of dephasing and excitation relaxation measurements in GaAs quantum wells using two pulse and three pulse photon echoes. Specifically important for determining the extent of spectral diffusion and localization and delocalization phenomena.

The general four-wave mixing configuration for frequency domain spectroscopy consists of two counter-propagating pump beams intersected in the resonant material by a probe beam at angle  $\theta$  (see Fig. 1). The beams are characterized by fields  $E_f(\omega_f, \vec{k}_f)$ ,  $E_b(\omega_b, \vec{k}_b)$ , and  $E_p(\omega_p, \vec{k}_p)$ , respectively. The fields interact in the medium to produce a coherent signal,  $E_s(\omega_s = \omega_f + \omega_b - \omega_p, \vec{k}_s)$ , through the nonlinear polarization of interest

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which is proportional to  $E_f E_b E_p^*$ . Spectroscopic information can be obtained by studying the response as a function of the different  $\omega$ 's, the angle  $\theta$ , the intensity, and the relative field polarizations. The nonlinear

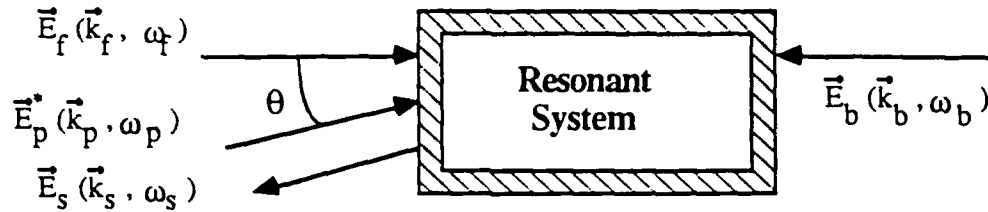


Figure 1. Geometry for backward four-wave mixing.

response is calculated using a quantum mechanical transport equation for the population density matrix, including relaxation:

$$i\hbar \left[ \frac{\partial}{\partial t} + \vec{v} \cdot \nabla \right] \rho = [H_o, \rho] + [V, \rho] - i\hbar \left. \frac{\partial \rho}{\partial t} \right|_{\text{relaxation}} + i\hbar A$$

where  $H_o$  is the internal Hamiltonian of the system,  $V$ , represents the electromagnetic interaction with the optical fields  $(-\vec{\mu} \cdot \vec{E})$ ,  $A$  describes incoherent pumping, and the relaxation term describes the coupling of the resonant system with the surrounding environment and includes not only decay out of  $\rho_{ij}$ , but also decay into  $\rho_{ij}$  from other states or coherences (for a review of relaxation, see Cohen-Tannoudji, 1977). The velocity term on the left hand side describes motion of the center of mass for gas phase systems and is usually ignored for crystals. The macroscopic polarization is given by the integral of  $\text{Tr}(\vec{\mu} \rho)$  where for resonances in gas phase systems, the integral is over velocity and for resonances in solids, the integral is over the distribution of resonant frequencies if the material is inhomogeneously broadened. This equation can be modified to include excitation diffusion effects in crystals, as we discuss below.

## Atomic Systems

In experiments where the pump beam frequencies are held fixed and the FWM response is measured as a function of the relative probe frequency (designated FWMp), our work in gas phase systems first demonstrated that nonconservation of population, alignment, or orientation due to spontaneous emission could lead to the presence of narrow resonances. In a more fundamental sense, these results showed the differences in the nonlinear spectral response between open and closed systems. For example, a system is closed with respect to population if the total population (upper state plus lower state) is conserved to within an overall decay constant.

The general form of the polarization is given by:

$$P = C \left| \frac{1}{[2\gamma_{12} - i(2\Delta - \delta)]} \right| \left| \frac{1}{(\gamma_2 - i\delta)} + \left\langle \frac{r}{\gamma_1 - i[\delta - (\mathbf{k}_p - \mathbf{k}_f) \cdot \mathbf{v}]} \right\rangle \right| \quad (1)$$

where  $\gamma_{12}$  is the natural linewidth (total coherence decay rate),  $\gamma_2$  is the upper state decay rate,  $\gamma_1$  is the ground state decay rate and  $\langle \rangle$  denotes velocity average.  $r$  is a parameter which is calculated based on the details of the relaxation and the energy levels of the system. If the system is closed,  $r=0$ . However, if the system is open,  $r$  can be negative or positive, resulting in either a dip or a spike in the nonlinear response.

In the simplest demonstration, Fig 2a shows the FWM response predicted for a closed Doppler broadened atomic two level system. The probe frequency is scanned with respect to the two constant pump frequencies, where  $\omega_f = \omega_b = \omega = \omega_0 - \Delta$  and  $\Delta$  is the pump resonance detuning. The peak on the left is actually the sum of two two-photon resonances: one has a width given by the upper state relaxation rate and the second has a width given by the ground state relaxation rate. The peak on the right is the sum of a one photon resonance and a three photon resonance and has a width given by twice the dipole dephasing rate. For a collisionless closed two level system, the amplitude of the ground state resonance is zero, and the widths of the two peaks are identical. However, in the presence of upper state radiative decay to a third level, say another hyperfine level in the ground state, the transition no longer conserves population. Now, the ground

state resonance contributes with a width given by the transit time, resulting in a near  $\delta$ -function type response, as shown by the solid line in Fig. 2b. However, there is a nonvanishing angle between the forward pump and probe, resulting in a residual Doppler width. The dashed line in Fig.

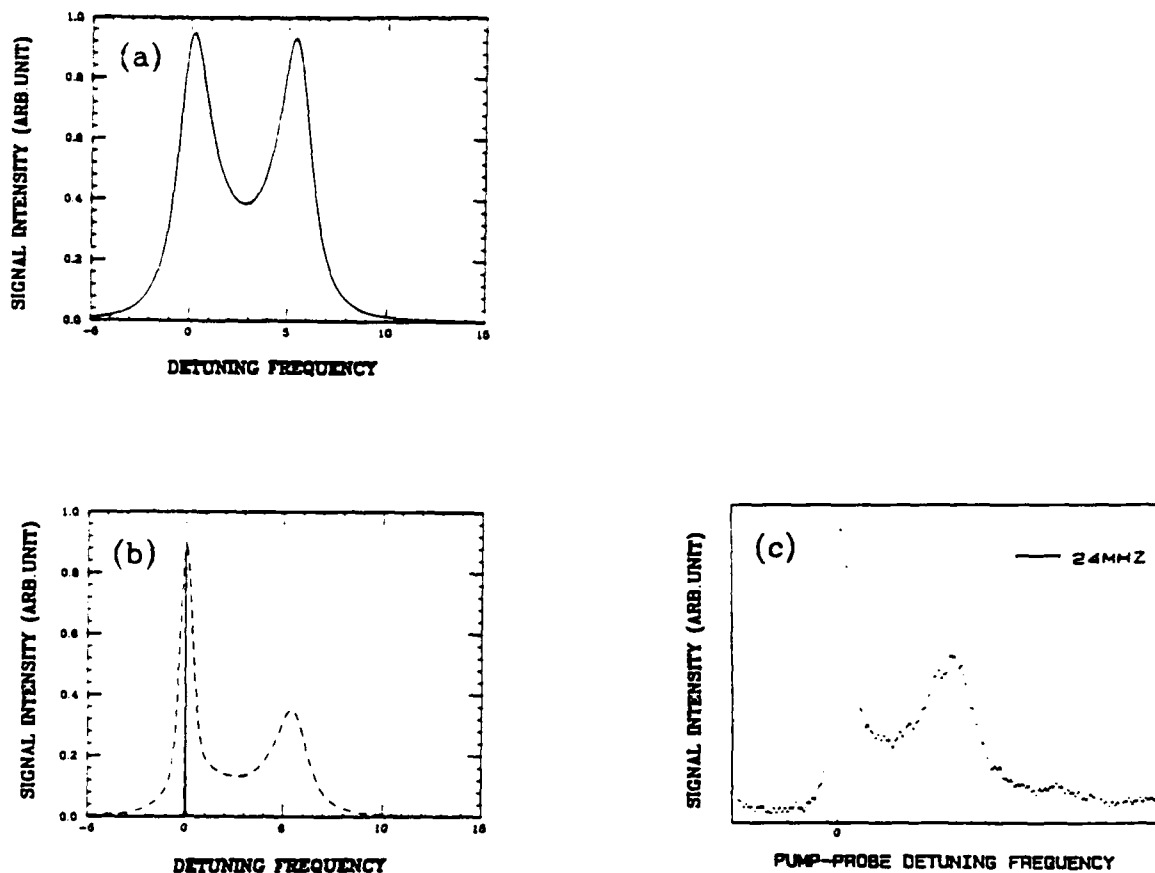


Fig. 2 (a) The theoretical FWM response for a closed two level system. (b) The theoretical FWM response for an open two level system. The dashed line includes the effects of residual Doppler broadening. (c) The measured spectral response.

2b shows the result. The narrow resonance still clearly persists. We note that in this case, the upper state decay rate exceeds the lower level decay rate resulting in a constructive interference (i.e.,  $r > 0$ ). The experiments were performed on the  $3s^2S_{1/2}(F=2)-3p^2P_{1/2}(F=2)$  transition in atomic sodium. This transition is open with respect to population since the upper

level can decay to the  $F=1$  ground state. Fig. 2c shows the experimental results which should be compared to the dashed line in Fig. 2b (see Liu, Remillard, Steel, 1987).

The effects of nonconservation of alignment and orientation were examined on a transition which is closed with respect to population. For these calculations, one must include magnetic degeneracies. In this case, we note that  $r$  can be positive or negative. In the case of the  $3s^2S_{1/2}(F=2)-3p^2P_{3/2}(F=3)$  transition, the upper state can decay only back to the  $F=2$  ground state. However, excitation of the transition with polarized light results in alignment of the excited state and ground state. The excited state alignment can decay to the ground state, further increasing the ground state alignment. In this case the  $r$  value is negative, resulting in a dip. Using cross polarized light, Fig. 3a and 3b show the measured and predicted response demonstrating very good agreement. (The theory includes coupling to the nearby excited state transitions, which does not significantly affect the predictions for the cross polarized geometry.) A similar argument describes the prediction for the co-polarized measurement, though the depth of the dip is not as deep. However, the experiment shows that a spike is observed. In this case, the contribution to the theory from the nearby transitions which do not conserve population is significant, and is critical to obtain agreement. Fig. 3c and 3d show the experiment and theory (see Berman, et. al. 1988).

The importance of this spectroscopy was shown in earlier work in  $\text{Cr:YAlO}_3$  where very narrow resonances were observed due to the presence of long lived metastable states (Steel and Rand, 1985). However, this work motivated an analysis of the high intensity response of this system. Using a semiclassical approach to the problem, we examined the high power limit. In the simple two-level problem, this leads to the prediction (Harter and Boyd, 1980) and observation (Steel and Lind, 1981) of the AC Stark effect. However, the results of this work show that in the presence of a long lived intermediate state, the FWM response broadens linearly with intensity (see Steel, Rand, Liu. 1987).

In the presence of collisions with ground state perturbers, the interactions becomes significantly more complex. The effects of collisions on such a system was described by Gorlicki, Berman and Khitrova (1988) where velocity changing effects on the upper and lower state population was described. We made a simple extension to the theory to include the effects

of state changing collisions. Using this model we compared theory and experiment using the  $3s^2S_{1/2}(F=2)-3p^2P_{3/2}(F=3)$  transition in atomic sodium

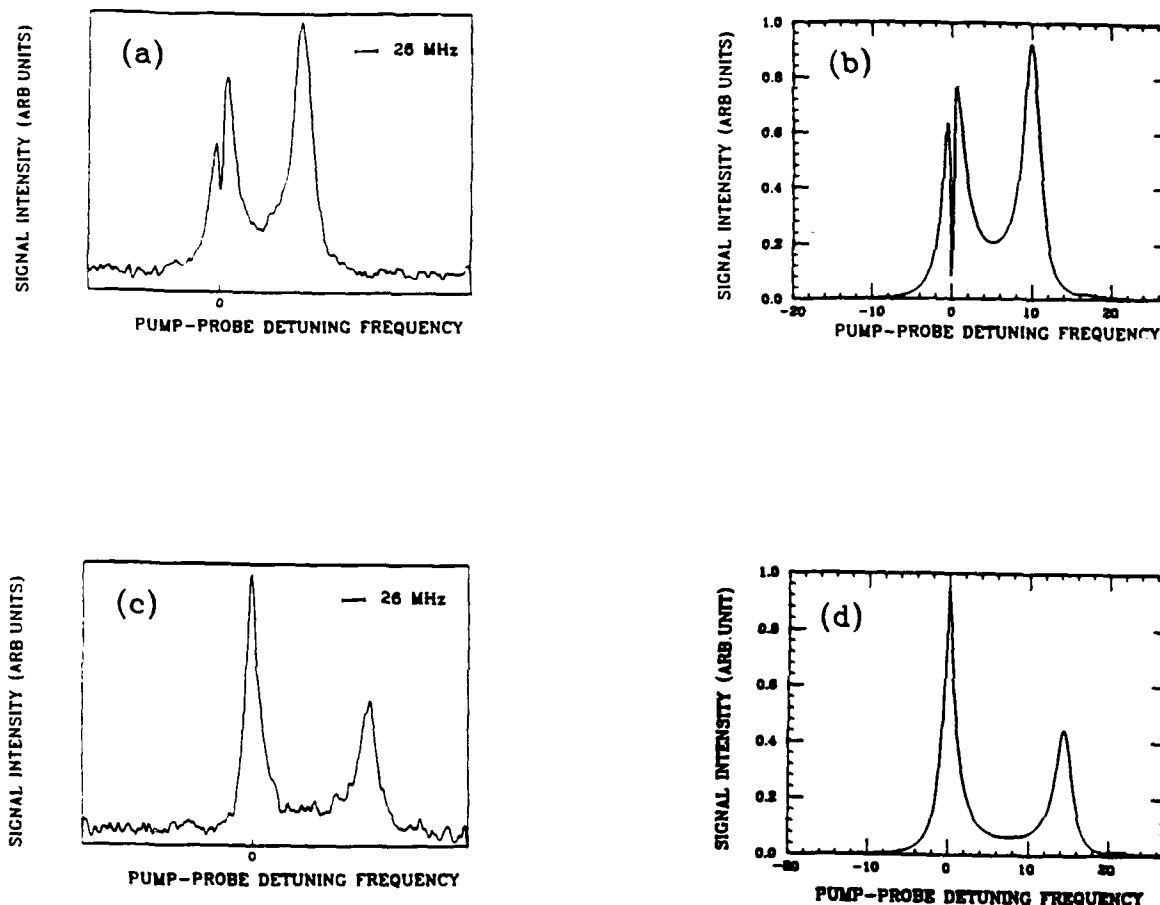


Figure 3. (a and b) Experiment and theory showing the presence of a negative dip in the cross-polarized FWM response caused by nonconservation of alignment and orientation. (c and d) Experiment and theory showing the spike that results in the co-polarized experiment. The dip becomes a spike because of the relative importance of population decay from nearby transitions.

in the presence of helium buffer gas. Figure 4 shows the results. In absence of collisions, Figure 4a and 4b show the dip due to nonconservation of alignment. At 5.6 Torr helium (4c and 4d), velocity changing collisions and state changing collisions result in nonconservation of population, and



a narrow spike is observed with a width given by the residual Doppler width due to the nonvanishing angle between the forward pump and probe. However, at high pressure (56 Torr., 4e and 4f), the principle feature has narrowed

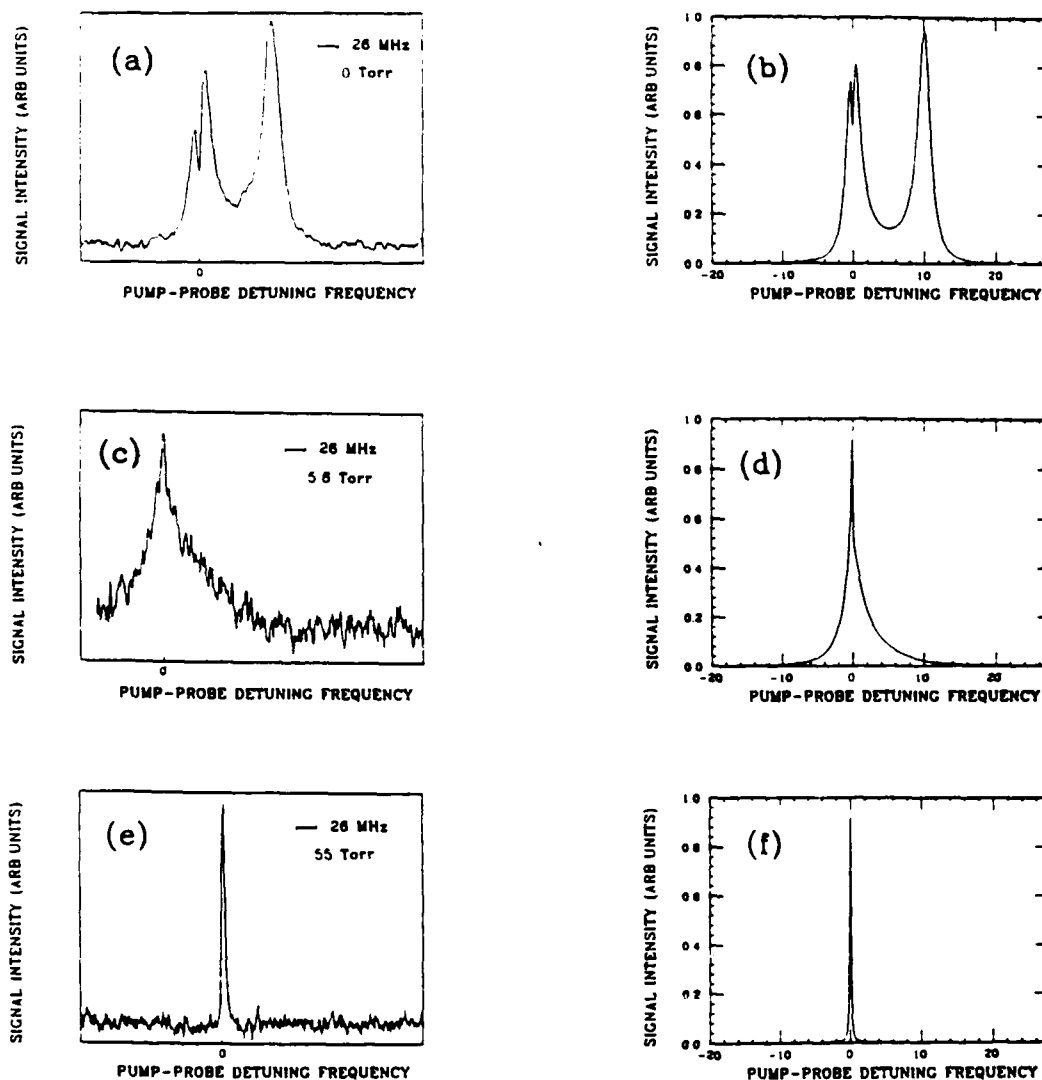


Figure 4: FWMp response in sodium in the presence of collisions. A comparison of theory and experiment. Note the last two figures show the first demonstration of Dicke narrowing on the spectral response of population dynamics.

below the residual Doppler width. This is a form of Dicke narrowing on the population dynamics. (see Liu and Steel, 1988).

The final work in atomic sodium resulted in a demonstration of FWM on Raman type transitions (V- and inverted V-transitions.) This work is important because it demonstrates a method to study relaxation associated with Raman coherence, a result difficult to obtain with other methods. The experiments show both excited state and ground state Raman coherence between hyperfine split states and magnetic substates. The results are being prepared for publication and are expected to be important for our work in semiconductor systems.

### Semiconductor Systems

The primary work in semiconductor heterostructures was in multiple quantum wells, discussed below. However, our first experiments were performed in glasses doped with semiconductor microcrystallites. These materials have been the subject of numerous recent studies because their large third order optical susceptibility is of interest for applications. In addition, it was believed that these materials might provide a simple material for the study of quantum size effects since the microcrystallite size can be in the range of  $40\text{\AA}$ . Our experiments have shown that the FWMp response obtained by tuning the probe beam is characterized by a narrow resonance. Based on our current work, we believe this resonance arises from a thermally activated trap. We have performed measurements as a function of temperature down to helium temperatures, and obtained a thermal activation energy of 120meV. Our additional spectroscopy studies including differential transmission suggest that the nonlinear response near the fundamental bandedge is most likely due to bandfilling (Remillard, et al, 1987, 1988a,b, Steel et al., 1988).

Most of our work in semiconductors was done in GaAs/AlGaAs multiple quantum wells (MQW) which are the subject of a large literature because of their importance to applications and condensed matter physics. Excitons dominate the linear and nonlinear optical properties of these structures near the fundamental band edge. Our study of the nonlinear optical properties has resulted in the first demonstration of high resolution spectroscopy of the excitons. In particular, we have obtained both room temperature and low temperature nonlinear optical spectra which provide new

insight into the structure and relaxation associated with these excitations. We briefly discuss the low temperature results.

As motivation for the work, we note the coupling of excitons to the applied radiation field is modified by dynamical interactions such as spontaneous emission, tunneling, diffusion, and scattering from phonons and defects. The processes can result in decay of the excitation as well as decay of the coherence or induced polarization between the initial and the excited state. Obtaining a quantitative description of the physics of relaxation is important since this understanding provides deeper insight into the fundamental properties of the material, and because relaxation affects the optical properties.

If the fluctuations are considered to give rise to small random shifts in the resonant frequency, designated  $\delta\omega_0(t)$ , then the decay of the induced polarization is given by  $\langle \exp[-i\int_0^t dt' \delta\omega(t')] \rangle$  where  $\langle \rangle$  denotes the ensemble average (see for example, Anderson, 1949, Breene, 1957). There are several methods for the evaluation of this term including the use of reservoir theory in the density matrix (Cohen-Tannoudji, 1977). For many cases of interest, the polarization decays exponentially with a resultant Lorentzian lineshape for the absorption profile. In general, however, the interaction is more complex and the lineshape may be non-Lorentzian. We designate the lineshape by  $f(\omega-\omega_0)$ , where  $\omega_0$  is the line center frequency. In a MQW, static fluctuations in the well thickness lead to energy level shifts of the exciton which vary with location. The resultant lineshape is then given by the average over these effects:  $F(\omega) = \int d\omega_0 P(\omega_0) f(\omega-\omega_0)$  where  $P(\omega_0)$  is a distribution function. If  $f$  and  $P$  are characterized by widths  $\Gamma$  and  $\Delta\omega_{inh}$ , respectively, the material is inhomogeneously broadened when  $\Delta\omega_{inh} \gg \Gamma$ . In this limit, linear absorption profiles provide little information on  $f$ . In a typical MQW at low temperatures, the excitonic features are inhomogeneously broadened since the linear absorption linewidth is of order 0.5-2 meV, while the intrinsic exciton linewidth may be an order of magnitude smaller.

This research describes the first direct experimental measurement of the lowest energy heavy hole exciton (HH1) lineshape (actually, the square of a function which is the lineshape convoluted with itself) in an inhomogeneously broadened MQW at low temperatures (see Remillard et al., 1988c,d). Using FWM, we have demonstrated that there are at least three

different states associated with the relaxation of the nonlinear response of the HH1 excitation. Specifically, we show that the three different states we have studied have lifetimes of order 100psec, 30nsec, and 10μsec, respectively. The lineshape,  $f(\omega-\omega_0)$  associated with the 100psec state is reasonably symmetric (i.e., nearly Lorentzian), however, the lineshape associated with the 16nsec state is highly asymmetric. *We note that these results cannot be completely described by the existing work on MQW excitons.*

In inhomogeneously broadened material for  $\vec{E}_b \perp \vec{E}_f \parallel \vec{E}_p$ , measurement of  $|E_s|^2$  as a function of  $\omega_p - \omega_f = \delta$ , the FWMp response (for  $\omega_f = \omega_b = \text{constant}$ ), gives a measure of the relaxation rates associated with the optical field induced perturbation of the excited states and the ground state, provided there is no orientation signal (i.e.,  $E_s = 0$  if  $\vec{E}_b \parallel \vec{E}_f \perp \vec{E}_p$ , which we have verified experimentally for low power cw excitation.) Moreover, for fixed  $\delta$ , measurement of  $|E_s|^2$  as a function of  $\omega_b - \omega_f$  ( $\omega_f$  constant) gives the square of the modulus of the convolution of  $f(\omega-\omega_0)$  with itself. We designate this the FWMb response (Steel and Remillard, 1987).

More specifically for the first case in a rate equation approximation, the equation of motion for  $\rho_{nn}$  (the probability per unit volume corresponding to the perturbation of state, n) may be solved to give:

$$\rho_{nn} = \frac{\zeta_n E_f E_p^*}{i\delta + \gamma_n + D_n |\Delta \vec{k}|^2} \exp(i\delta t + i\Delta \vec{k} \cdot \vec{x}) + \text{c.c.} \quad (2)$$

where  $\gamma_n$  is the exponential decay rate,  $D_n$  is the spatial diffusion coefficient,  $|\Delta \vec{k}|^2 = |\vec{k}_f - \vec{k}_p|^2 = (2k)^2 \sin^2(\theta_{\text{int}}/2)$ ,  $\theta_{\text{int}}$  is the internal angle between the forward pump and probe and we have taken  $k_f \cong k_p = k$ . (We have not included the effects of source terms in the above equation due to reservoir effects and we have assumed the dephasing rate is much larger than state relaxation rates. Corrections to these assumptions can lead to a significant modification of the FWMp spectral response (Remillard and Steel, 1987, Berman, Steel, Ghitrova, Liu, 1988.) We see then that tuning  $\delta$  provides information on the relaxation of these states. Furthermore, the lineshape dependence on  $D_n$  and  $\theta$  gives a way to measure  $D_n$  (Remillard, et al., 1988e). There is a corresponding method in the time domain which has been used to study exciton localization effects in these materials

(Hegarty, et al, 1984a).

In the second case,  $\omega_b$  is scanned while  $\delta$  is fixed, yielding the FWMb response. If  $\delta$  is small compared to a specific  $\gamma_n$  [and hence much smaller than the characteristic width of  $f(\omega-\omega_0)$ ], then the forward pump and probe produce a spatial modulation of the states characterized by resonant frequencies  $\omega_0'$  around  $\omega$  ( $\approx \omega_f \approx \omega_p$ ). If  $N(\omega_0')$  represents the difference between the upper level and lower level excitation, we see that  $N(\omega_0')$  varies as the mirror image of  $f$ , i.e.,  $f(\omega_0'-\omega)$ . The lineshape of the resonant group probed by the back pump is  $f(\omega_b-\omega_0')$ , and this leads immediately to the FWMb spectral response measured by the signal intensity:

$$I = K \left| \int dx P(\omega-x) f(x) f(\Delta-x) \right|^2$$
 where  $x = \omega - \omega_0$ ,  $\Delta = \omega - \omega_b$  and  $K = K(\delta)$  contains Lorentzian denominators associated with terms like Eq. (2). We see that if we are tuned near the center of the distribution function  $P$  (assumed to be broad), the signal response is the square of the convolution function of the lineshape with itself. Furthermore, if there are different transitions characterized by different  $\gamma_n$ 's and corresponding lineshapes, controlling  $\delta$  provides a means of measuring the different lineshapes.

The above discussion is *qualitative* and based on detailed understanding of relaxation effects in simpler systems using the density matrix formulation. Even for these systems, the nonlinear response is often more complex because it is the square of the sum of different time order perturbation sequences. However, most of these problems do not contribute to these experiments, since no orientation signal is detected and the characteristic width of  $f$  is greater than specific state relaxation rates. A more rigorous description and deeper physical insight should be possible based on the recent development of effective optical Bloch equations for semiconductors (Lindberg and Koch, 1988 and included references), but such an analysis would not be expected to change the qualitative understanding.

The linear absorption spectrum shows the clearly resolved HH1 and the lowest light hole (LH1) excitons, with each peak having a width less than 2 meV. For degenerate FWM (DFWM), the pump intensity is of order 600 mW/cm<sup>2</sup> corresponding to an exciton density in the well region of  $5 \times 10^6$  cm<sup>-2</sup>/layer. The DFWM response shows only one peak, shifted 20 cm<sup>-1</sup> to the red with respect to HH1, and is most likely associated with the heavy hole localized at island defects (Masumoto, et al., 1984, Takagahara, 1985).

Studies of LH1 will require the use of a tighter focusing geometry and have been deferred to later work (see Section III).

The FWMp response is shown in Fig. 5. Figure 5a shows two resonances with linewidths of order 3 GHz and 10 MHz, respectively (the narrow feature was measured with increased dispersion.) A scan of the tip of Fig. 5a was obtained using the method of correlated optical fields to eliminate contributions from interlaser jitter (3MHz). Figure 5b shows the corresponding spectrum. A Lorentzian fitted to the tip corresponds to a linewidth of the order of 30kHz. From Eq. (2), we see that the broad component in Fig. 5a corresponds to a lifetime of 100 psec while the narrow feature shows a lifetime of 30 nsec. In Fig. 5b, the lifetime is of order 10 $\mu$ sec. We ascribe the fast time to the intrinsic

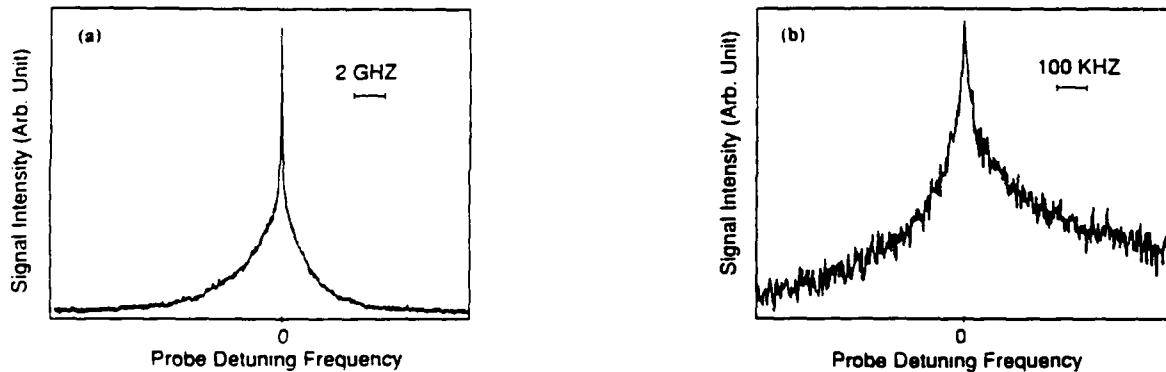


Figure 5. The FWMp response in a GaAs/AlGaAs MQW at 5K. The data shows that there are three different decay rates associated with HH1.

lifetime of the exciton. The value is comparable to the most recent measurements (Honold, et al., 1988) performed at similar intensities. The origin of the longer lifetimes and the nature of the associated states are not clear at this time. A possible explanation for the origin of the 30

nsec state is that local electric fields resulting from surface or bulk localized states may result in a splitting of the electron and hole, an effect which is known to increase the exciton lifetime due to the reduced overlap of the wavefunctions (Pollard, et al., 1985). The lineshape data shown below indicates that these longer lived states may play an important role in the interpretation of earlier work. (The slight asymmetry in Fig 5a is possibly the result of interference effects in the nonlinear response and will be studied using a modified FWMp approach discussed below.)

Using the above information, we measured  $f(\omega - \omega_0)$  for  $\delta = 40$  MHz (to reduce contributions from the longer lived states,) and for  $\delta = 500$  kHz, (to include contributions from the 16nsec lifetime state.) Figure 6a shows the FWMb spectrum recorded

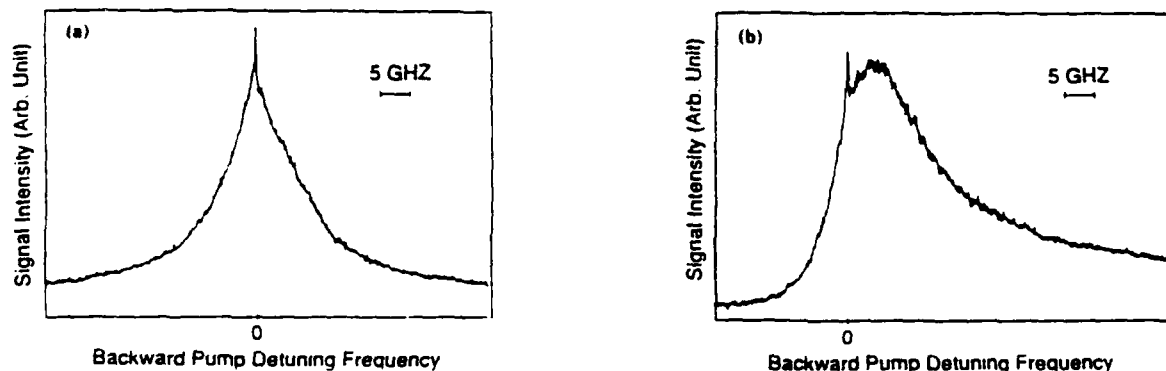


Figure 6. The FWMb response in a GaAs/AlGaAs MQW at 5K. This data provides a direct measurement of exciton lineshape. (a) shows the lineshape on the low frequency side of the nonlinear response with  $\delta = 40$  MHz, (b) shows the lineshape at the peak of the absorption with  $\delta = 500$  kHz.

for  $\delta = 40$  MHz (the spike may be due to polarization leakage from the stress induced birefringence in the cryostat windows and is ignored in the present discussion.) A Lorentzian curve fits the wings of the data quite well in Fig. 6a, with some deviation on the high frequency side. Ignoring this deviation and using the relationship  $\gamma_{ph}^{-1} = 1/2\pi\Delta\nu$ , the decay time is 46psec.

A time of 32psec has been measured in this sample independently using a three pulse (stimulated) photon echo (discussed below.) The deviation from a Lorentzian suggests that the line broadening mechanism is more complex than a simple exponential decay of the polarization. This effect becomes much more dramatic if we examine the lineshape with  $\delta=500\text{kHz}$  to increase the contribution from the longer lived states. On the red side of the nonlinear response, the lineshape is similar to Fig. 6a, however, on the blue side of the response (approaching absorption line center), the lineshape is shown in Fig. 6b. The profile exhibits a high degree of asymmetry and an effective increase in linewidth. Such an increase in the linewidth was observed earlier in resonant Raleigh scattering (Hegarty et al., 1982a) and attributed to the onset of delocalization. The linewidth changes which we observe are consistent with the earlier report, however, *we note that this change is due to contributions from the longer lived states.*

The origin of the lineshape will likely come from a correct application of the scattering terms in the effective optical Bloch equations (Lindberg and Koch, 1988 and included references), and not just simple curve fitting of multiple Lorentzians. However, some physical insight into this behavior may be had by considering the quasi-static line broadening theory [see the generalized discussion by Anderson (1948)] which shows that long range static or slowly varying interactions cause shifts in the levels which contribute to the lineshape. Under certain conditions, these shifts can lead to asymmetry in the line. If the interactions are through an attractive electrostatic coupling, the asymmetry is to the red while if the potential is repulsive, the asymmetry is to the blue. We are currently determining the effect of quasistatic line broadening effects on FWM. An alternative explanation which we are pursuing is that there may be relaxation due to time dependent changes in the resonant frequency (Berman, 1986a,b), due to effects such as tunneling or variable range hopping. Such a calculation would also give rise to asymmetric lineshapes in the FWMb response. We are currently exploring methods to distinguish these different possibilities.

Our most recent work involves the demonstration and use of three pulse photon echos. We were particularly interested in these measurements to permit the comparison between time domain and frequency domain measurements



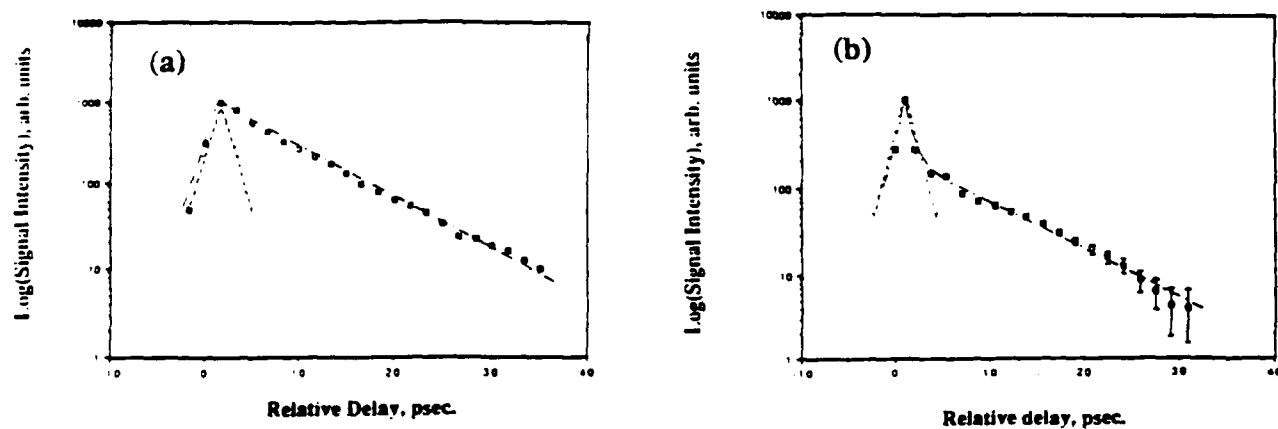


Figure 7. Stimulated photon echo response as a function of forward pump probe delay. (a)  $16\text{cm}^{-1}$  detuned to the red with zero time delay in the back pump beam. (b)  $16\text{cm}^{-1}$  detuned to the blue with a 40psec delay in the back pump beam.

of relaxation. There are at least two important reasons why these results would differ: the photon flux associated with short pulse measurements could lead to density effects; spectral diffusion may dominate dephasing on the long time scales of cw frequency domain measurements. It is extremely interesting to note that at the peak of the nonlinear response, two pulse and three pulse photon echo measurements give a dephasing time of 32 psec and frequency domain measurements give 46 psec (assuming a Lorentzian form). We consider this to be a reasonable comparison since the frequency domain results show the profile is not completely Lorentzian. However, we show that as the excitation wavelength decreases, the echo measurements show the dephasing rate increases. Moreover, if we delay the third pulse 40 psec, we observe an extremely fast initial decay, limited by our pulse width. (See Figure 7) In a three pulse echo (stimulated photon echo), the dependence of dephasing rate on the delay of the third pulse is evidence of spectral diffusion. Besides extending our work in this area, we are

concentrating on making the first clear demonstration of delay in the echo signal and eliminating other possible explanations for the above behavior. Further work in this area will be described in Section III.

## **ARO Publication Summary and Student Progress**

### **Publications supported by this program**

1. J.T. Remillard, H. Wang, D.G. Steel, "High Resolution Nonlinear Laser Spectroscopy of the Heavy Hole Exciton in a GaAs/AlGaAs Multiple Quantum Well: A Direct Measure of the Exciton Lineshape," submitted to Phys. Rev. Lett.
2. H. Wang, J. Liu, D.G. Steel, "Nonlinear Raman Effects in the Four-Wave Mixing Spectrum of Hyperfine Structure and Magnetic Substates in Atomic Sodium," to be submitted to Phys. Rev. A.
3. J. Liu and D.G. Steel, "High Resolution Four-Wave Mixing Spectroscopy of Collision Induced Narrow Resonances in Doppler Broadened Systems", accepted for publication in Physical Review A, 1988.
4. P.R. Berman, D.G. Steel, G. Khitrova, and J. Liu, "Effects of radiative decay in four-wave mixing spectroscopy: narrow resonances produced by nonconservation of population, alignment and orientation," Phys. Rev. A 38, pp252-262 (1988).
5. D. G. Steel, J. T. Remillard, Jing Liu and S. C. Rand, "Effects of population pulsations on backward nearly degenerate four-wave mixing spectroscopy and optical phase conjugation," Jr. Opt. Soc. Am. B, 5, pp171-179 (1988).
6. Jing Liu, J. T. Remillard and D. G. Steel, "Decay induced narrow resonances in backward nearly degenerate four-wave mixing spectroscopy," Phys. Rev. Lett. 59, pp779-782 (1987).
7. D.G. Steel, S.C. Rand, Jing Liu, "Four-wave mixing studies in Cr:YAlO<sub>3</sub> for application to optical phase conjugation," J. Opt. Soc. Am. B 4, pp1794-1800 (1987).
8. J. T. Remillard and D. G. Steel, "Nonlinear laser spectroscopy studies of semiconductor materials," Laser Spectroscopy VIII, Springer-Verlag, W. Persson and S. Svanberg, eds., pp264-266 (1987).
9. Jing Liu, Galina Khitrova, Duncan Steel and Paul Berman, "Narrow Resonances in Four-Wave Mixing Due to Radiative Decay", Laser Spectroscopy VIII, Springer-Verlag, W. Persson and S. Svanberg, eds., pp291-293 (1987).
10. D.G. Steel, "High Resolution Nonlinear Laser Spectroscopy of GaAs/AlGaAs Multiple Quantum Wells." invited, to be presented at the OSA

Topical Meeting on Quantum Wells for Optics and Optoelectronics, Salt Lake City, March, 1989.

11. J.T. Remillard, H. Wang, M.D. Webb, and D.G. Steel, "Frequency domain four-wave mixing spectroscopy in semiconductor quantum wells and microcrystallites," XVI International Conference on Quantum Electronics, Technical Digest, pp278-279, IQEC Tokyo (1988).

12. Jing Liu, D.G. Steel, P.B. Berman, M. Gorlicki, and G. Khitrova, "High resolution four-wave mixing spectroscopy of narrow resonances in Doppler broadened systems," XVI International Conference on Quantum Electronics, Technical Digest, pp36-37, IQEC Tokyo (1988).

13. J.T. Remillard, Hailin Wang, D.G. Steel, "High Resolution Nonlinear Laser Spectroscopy of the Heavy Hole Exciton at 5 K: A Direct Measure of the Exciton Lineshape," XVI International Conference on Quantum Electronics, Technical Digest (Postdeadline), pp48-49, IQEC Tokyo (1988).

14. D.G. Steel, J.T. Remillard, H. Wang, and M.D. Webb, "Application of 4-wave mixing spectroscopy to semiconductor material studies", invited, Conference on Lasers and Electro-Optics, Technical Digest Series, 7 pp260-261 (1988).

15. D.G. Steel, Jing Liu, P.R. Berman, and G. Khitrova, "Radiative decay induced narrow resonances in 4-wave mixing due to nonconservation of population, alignment and orientation", Conference on Lasers and Electro-Optics, Technical Digest Series, 7 pp186-187 (1988).

16. J.T. Remillard, J. Liu, and D.G. Steel, "Application of frequency domain 4-wave mixing spectroscopy to semiconductors: studies of semiconductor doped glasses," Proceedings of the International Workshop on High-Speed optical processes and opto-electronic devices based on compound semiconductors, p F9, National Science Foundation Workshop, The University of Michigan, 1987.

17. Duncan Steel, Jing Liu, Paul Berman, and Galina Khitrova, "Narrow resonances in four-wave mixing due to radiative decay", International Laser Science Conference, Atlantic City, 1987.

18. D. G. Steel, S. C. Rand, and J. Remillard, "Ultrannarrow optical resonances and measurements of relaxation rates using resonant FWM," International Conference on Quantum Electronics 1986.

### Student Progress Summary

Four students have been supported in this program. All of them have reached Ph.D. candidacy status. One student received the Ph.D. degree in early 1989 and a second student will receive his degree in early fall. A fifth student will join the program shortly. The program draws on students from the Departments of Physics and Electrical Engineering and the Applied physics program.

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